REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YYYY)*	2. REPORT TYPE	3. DATES COVERED *(From - To)*
APRIL 2014	Journal Article (Post Print)	SEP 2012 – DEC 2013

4. TITLE AND SUBTITLE

DIRECT DETECTION OF TIME-RESOLVED RABI OSCILLATIONS IN A SINGLE QUANTUM DOT VIA RESONANCE FLUORESCENCE

5a. CONTRACT NUMBER
FA8750-12-2-0333

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)

J. Schaibley, A. Burgers, G. McCracken, D. Steel, A. Bracker, D. Gammon, and I. Sham

5d. PROJECT NUMBER
QEST

5e. TASK NUMBER
MI

5f. WORK UNIT NUMBER
CH

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
University of Michigan
450 Church Street
Ann Arbor MI 48109-1040

8. PERFORMING ORGANIZATION REPORT NUMBER
N/A

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Air Force Research Laboratory/Information Directorate
Rome Research Site/RITA
525 Brooks Road
Rome NY 13441-4505

10. SPONSOR/MONITOR'S ACRONYM(S)
AFRL/RI

11. SPONSORING/MONITORING AGENCY REPORT NUMBER
AFRL-RI-RS-TP-2014-026

12. DISTRIBUTION AVAILABILITY STATEMENT
Distribution Approved For Public Release; Distribution Unlimited. This report is the result of contracted fundamental research deemed exempt from public affairs security and policy review in accordance with SAF/AQR memorandum dated 10 Dec 08 and AFRL/CA policy clarification memorandum dated 16 Jan 09.

13. SUPPLEMENTARY NOTES

14. ABSTRACT

OpticalRabi oscillations are coherent population oscillations of a two-level system coupled by an electric dipole transition when driven by a strong nearly resonant optical field. In quantum dot structures, these measurements have typically been performed as a function of the total pulse area \int \Omega_0 dt where the pulse area varies as a function of Rabi frequency. Here, we report direct detection of the time-resolved coherent transient response of the resonance fluorescence to measure the time evolution of the optical Rabi oscillations in a single charged InAs quantum dot. We extract a decoherence rate consistent with the limit from the excited state lifetime.

15. SUBJECT TERMS
Quantum information processing, quantum entanglement, quantum computing

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			PAUL ALSING
U	U	U	UU	6	19b. TELEPHONE NUMBER (*Include area code*) N/A

Standard Form 298 (Rev. 8-98)
Prescribed by ANSI Std. Z39.18

Direct detection of time-resolved Rabi oscillations in a single quantum dot via resonance fluorescence

J. R. Schaibley, A. P. Burgers, G. A. McCracken, and D. G. Steel*
The H. M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan 48109-1040, USA

A. S. Bracker and D. Gammon
The Naval Research Laboratory, Washington, DC 20375, USA

L. J. Sham
Department of Physics, The University of California, San Diego, La Jolla, California 92093-0319, USA
(Received 13 November 2012; revised manuscript received 14 February 2013; published 19 March 2013)

Optical Rabi oscillations are coherent population oscillations of a two-level system coupled by an electric dipole transition when driven by a strong nearly resonant optical field. In quantum dot structures, these measurements have typically been performed as a function of the total pulse area $\int \Omega_0(t)\,dt$ where the pulse area varies as a function of Rabi frequency. Here, we report direct detection of the time-resolved coherent transient response of the resonance fluorescence to measure the time evolution of the optical Rabi oscillations in a single charged InAs quantum dot. We extract a decoherence rate consistent with the limit from the excited state lifetime.

DOI: 10.1103/PhysRevB.87.115311 PACS number(s): 81.07.Ta, 42.50.Hz, 42.50.Ar

I. INTRODUCTION

Optical Rabi oscillations have been observed in atomic and solid state systems and are essential for many coherent control protocols.[1–4] In semiconductor quantum dots (QDs), the structure of these oscillations gives insight into the time domain response and coherence properties when driven by a strong field. Here, we present a novel technique to directly observe optical Rabi oscillations by monitoring time-dependent resonance fluorescence from a single InAs QD. Due to experimental difficulties and the desire to use Rabi oscillations to manipulate the state rapidly, experiments demonstrating Rabi oscillations are usually performed with short pulses of fixed pulse width. In contrast to previous work where Rabi oscillations are observed by varying the power of the driving pulse and measuring either time integrated fluorescence or differential transmission,[1,2,4] we time resolve the single QD fluorescence under square pulse excitation, allowing for direct observation of the excited state population oscillations.[5] Our method allows for fast collection of high-quality data that can be used to characterize a QD transition's optical decoherence rate, oscillator strength, and can be used in quantum information processing as a spin qubit readout channel.

It is well known that the first- and second-order correlation functions of light scattered from a single two-level system, when driven by a continuous-wave (CW) field, will exhibit oscillations at the Rabi frequency.[6,7] These oscillations were observed most recently in a single InAs QD system by using standard intensity correlation (g^2) techniques.[8,9] We report a quasi-CW technique that allows for the Rabi oscillations to be controlled by the amplitude, width, and delay of the square driving pulse. Though this technique is used here to study a single QD transition, it can be applied to an ensemble of identical optical transitions, since time-zero is determined by the excitation pulse. The effect of CW Rabi oscillations has also been used to induce geometric phases in a single QD spin qubit system, which were observed by monitoring the effect of electron spin precession in the presence of a magnetic field.[10]

The basic theory of a two-level atom starting in the ground state and driven by a strong resonant CW laser yields the well-known Rabi solution, which predicts oscillation of the excited state population ρ_{22},

$$\rho_{22}(t) = \sin^2\left(\frac{\Omega_0 t}{2}\right), \tag{1}$$

where the Rabi frequency is given by $\Omega_0 = \mu E_0/\hbar$. μ is the dipole moment of the optical transition, and E_0 is the driving field amplitude. By solving the density matrix equations for a resonant driving field, including the effects of spontaneous emission and decoherence, the probability of being in the excited state as a function of time is given by

$$\rho_{22}(t) = \frac{\Omega_0^2/2}{\Omega_0^2 + \gamma\gamma_2} \times \left\{1 - \left[\cos(\lambda t) + \frac{\gamma+\gamma_2}{2\lambda}\sin(\lambda t)\right]e^{-\frac{1}{2}(\gamma+\gamma_2)t}\right\}, \tag{2}$$

where

$$\lambda = \sqrt{\Omega_0^2 - \frac{(\gamma-\gamma_2)^2}{4}}, \tag{3}$$

where γ_2 is the excited state decay rate, and γ is the decoherence rate. Equation (2) shows that the damping rate of the oscillation is the average of the excited state decay rate and the decoherence rate. Although this result is well known,[5,11] it is usually challenging to observe due to experimental limitations.

II. SAMPLE AND METHODS

The experiment is performed on a single negatively charged InAs QD, which has been shown to form an effective two-level system even under strong laser excitation.[12–15] The QDs are grown in a Schottky diode heterostructure by molecular beam epitaxy. The heterostructure consists of n-doped GaAs substrate, 40-nm intrinsic GaAs spacer layer, 2.8-nm InAs,

280-nm intrinsic GaAs capping layer, and a 5-nm Ti layer forming a Schottky diode. The sample is held at 5.5 K in a liquid helium optical cryostat. Biasing the Schottky diode enables deterministic charging of the QD with single carriers.[12,14] A singly negatively charged QD can be optically excited forming a trion state (i.e., a negative exciton). The trion state is identified by analyzing bias-dependent photoluminescence spectra following established work.[12,14,16] The charge state is then verified by performing polarization-dependent high-resolution voltage modulation absorption spectroscopy,[13,17] and CW resonance fluorescence.[8,18,19] The ground state consists of the degenerate spin eigenstates of a single electron in the lowest conduction band level with angular momentum projection on the growth axis (001) to be $|\pm\frac{1}{2}\rangle$. The lowest optically excited state consists of a pair of degenerate heavy-hole charged exciton (trion) states with angular momentum $|\pm\frac{3}{2}\rangle$. The two optically allowed electric dipole transitions are degenerate and are coupled by $\sigma+$ and $\sigma-$ polarized light. By exciting the QD with horizontally polarized light, we equally couple the degenerate transitions. We collect light emitted from the QD in the transmission geometry, and by using a polarization analyzer set to pass vertically polarized light, we can isolate the fluorescence emission of the QD from the excitation laser.

The excitation pulses are generated from a narrow bandwidth CW Ti:sapphire laser with sub-MHz linewidth. A waveguide electro-optic modulator and high-speed pulse generator are used to generate square pulses of controllable width with a pulse contrast of at least 25 dB. For the 2-ns square pulses used to observe the CW Rabi oscillations, the pulse rise time is approximately 200 ps. The excitation laser is passed through a linear polarizer and Babinet Soleil compensator before it is focused on the Ti side of the sample by a 0.68 NA aspheric objective lens. The QD resonance fluorescence is collected on axis in the transmission geometry by another 0.68 NA lens. Similar to other recent work,[18,20] the collected light is then passed through a crossed linear polarizer before it is coupled to a single mode fiber. The polarizer and fiber rejects nearly all the resonant excitation laser while passing half of the QD fluorescence. In this setup, we are able to achieve polarization extinction ratio exceeding 10^6. The light emitted from the QD is detected by a fiber coupled Geiger mode single photon avalanche detector (SPAD). The signal is recorded with a time-to-digital converter which is synchronized with the excitation pulse. The system's timing resolution is limited by the timing jitter in the detector to <48 ps FWHM. The fluorescence photons are time histogrammed over an integration time of two minutes at an experimental repetition rate of 76 MHz. The data are background subtracted and deconvolved from the system's instrument response function using a linear deconvolution technique.[21] The deconvolved data are fit to Eq. (2) to extract the Rabi frequency and decoherence rate by using the measured lifetime. The fits are then convolved with the instrument response function and plotted with the raw data.

III. RESULTS

A single Lorentzian is observed in the (low power) fluorescence spectrum with a linewidth of 623 ± 25 MHz (see Fig. 1). In order to observe the time-resolved emission process, we perform a time correlated single photon counting measurement under pulsed laser excitation. By using a high-speed electro-optic modulator, we gate the resonant CW laser in time and generate square pulses of controllable width and repetition rate. We generate 250-ps pulses that are shorter than the excited lifetime and observe the expected single exponential decay signal (see Fig. 2). As we increase the power of the pulse, we are able to observe the onset of power-dependent Rabi oscillations for a fixed pulse width of 250 ps (see Fig. 3 and inset).

Increasing the excitation pulse width to 2 ns, we are able to observe time-dependent Rabi oscillations of the excited state population (see Figs. 4 and 5). As expected, the oscillation

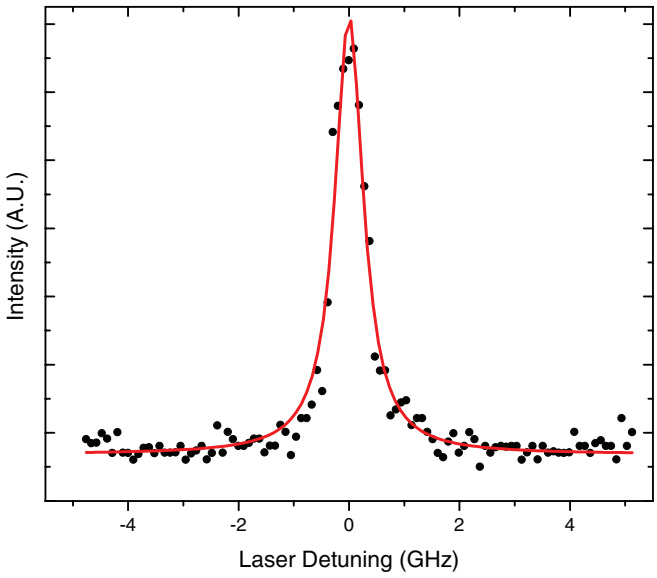

FIG. 1. (Color online) Single trion state fluorescence spectrum under resonant narrow bandwidth CW excitation. The red line is a Lorentzian fit to the data yielding a linewidth of 623 ± 25 MHz FWHM. The resonance energy is approximately 1.307 eV.

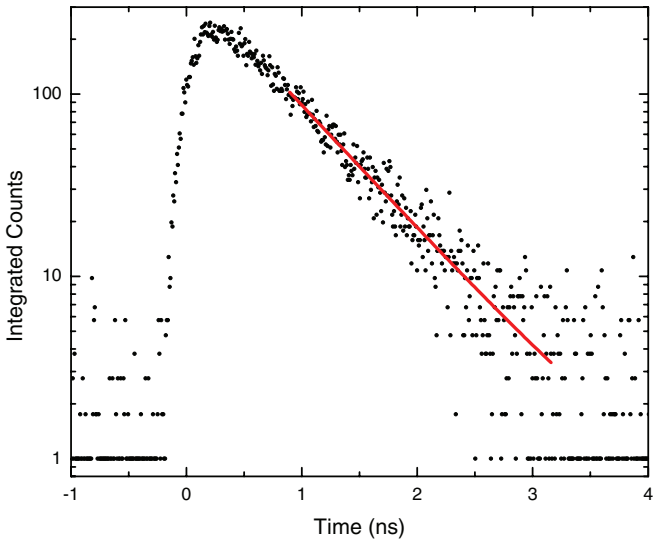

FIG. 2. (Color online) Time-resolved QD fluorescence under resonant 250 ps pulsed excitation. The red line is a single exponential fit to the data yielding a trion lifetime of 640 ± 25 ps.

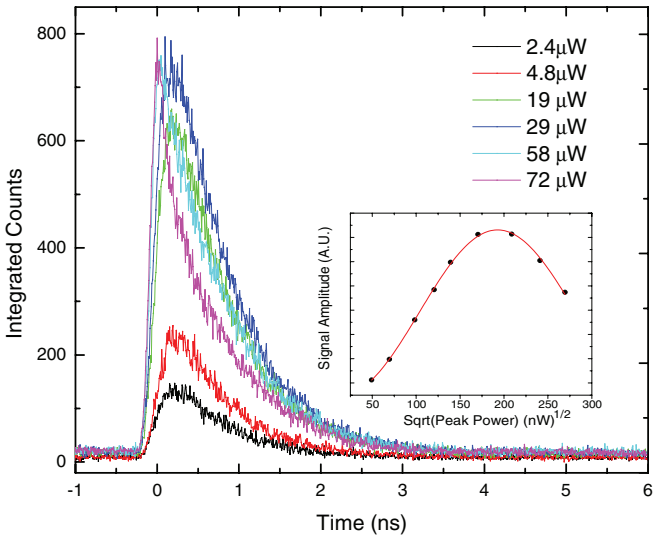

FIG. 3. (Color online) Power dependence of QD fluorescence under resonant 250 ps pulsed excitation. By extracting the amplitude of the time-resolved fluorescence after the pulse, we observe the onset of power-dependent Rabi oscillations shown in the inset. The highest power is limited by photorefractive effects in the electro-optic modulator used to generate the optical pulses.

frequency scales with the square root of the excitation power (see Figs. 5 and 6), and the Rabi oscillations are damped as the two-level system approaches steady state (equal probability of being in the ground and excited states). Though well known, it is somewhat non-intuitive that the damping rate of the Rabi oscillations goes as $\frac{1}{2}(\gamma + \gamma_2)$,[5,11] i.e., the average of the

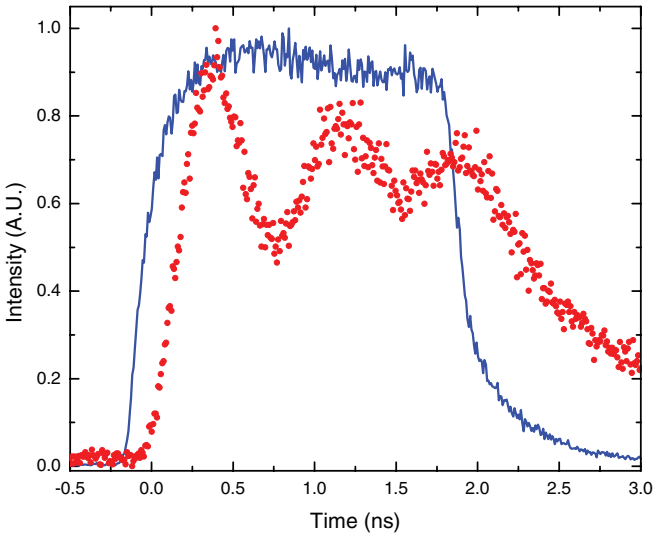

FIG. 4. (Color online) Normalized QD fluorescence (red) showing time-dependent Rabi oscillations while driven by a 10.2 μW peak power laser pulse. By leaking a small amount of the excitation laser onto the SPAD, we obtain a real-time measurement of the 2-ns driving pulse (blue), which is normalized and overlaid with a CW Rabi signal. Both the CW Rabi and pulse measurements are distorted by convolution with the detection systems's (asymmetric) instrument response function, which leads to an exaggerated rise/fall time of the square 2-ns pulse.

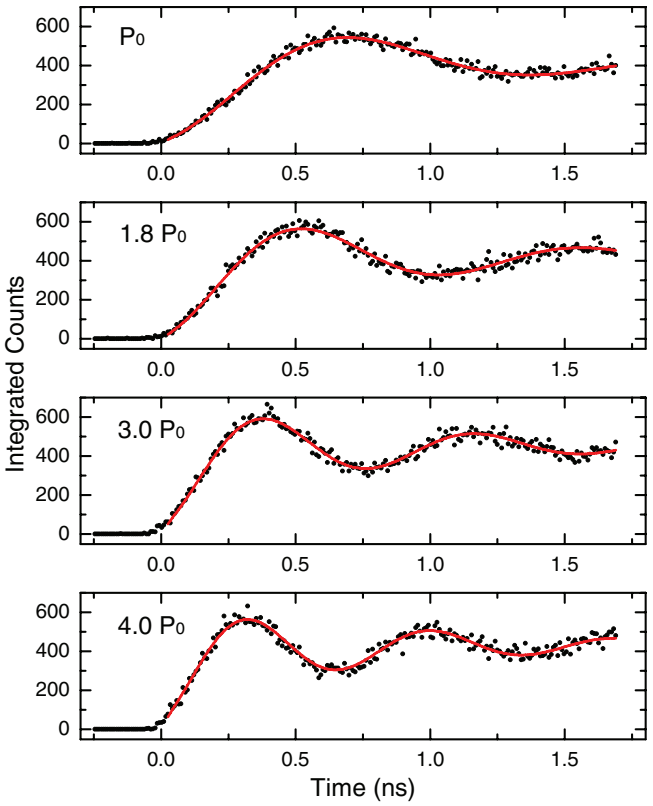

FIG. 5. (Color online) Time-dependent Rabi oscillations under 2 ns square pulsed excitation for increasing excitation powers. The lowest (peak) excitation power is $P_0 \approx 3.4\ \mu$W. The solid lines are convolved fits of Eq. (2) to the data.

transverse, γ, and longitudinal, γ_2, decay rates. The physical origin of this is that during the pulse, the optical Bloch vector, which is rotating around the x-axis, spends equal time oriented along both the z and y axes, decaying at the population decay rate, γ_2, and the decoherence rate, γ, respectively.

Using the trion lifetime extracted from the short pulse measurements (640 ± 25 ps), we are able to fit Eq. (2) to the data with only the Rabi frequency, Ω_0, and the decoherence rate, γ as physical fitting parameters. These data allow us to extract a transverse lifetime (i.e., the induced optical dipole decoherence time) of $1/\gamma = 1.22 \pm 0.06$ ns, which is consistent with the theoretical prediction of no pure dephasing where $\gamma = \gamma_2/2$. The ratio of γ to γ_2 is in agreement with previous frequency domain studies on single InAs QDs.[22] The γ values extracted from the Rabi fits appear randomly distributed about the mean, indicating no excitation induced dephasing effects (see Fig. 6).

IV. DISCUSSION

We note that the transverse lifetime of 1.22 ns extracted from the Rabi oscillation data is roughly two times longer than expected from the CW resonance fluorescence linewidth of 623 ± 25 MHz, which would yield $1/\gamma = 1/(\pi 623\text{MHz}) = 511 \pm 21$ ps. Typical linewidths of ≈600 MHz have been observed by several groups studying single InAs QDs,[8,15,18] and are believed to be broadened beyond the natural linewidth (2γ) due to a spectral wandering process.[15] It is promising that even

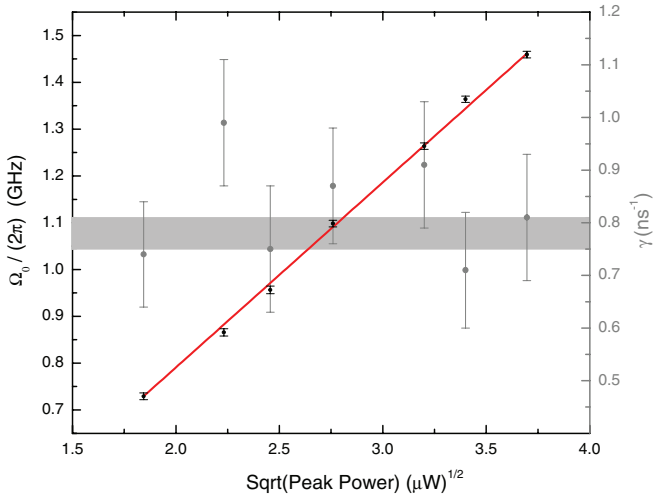

FIG. 6. (Color online) Rabi frequency (black) from time-resolved data plotted as a function of the square root of the excitation power showing the expected linear relationship. The red line is a linear fit to the data with the y intercept set to zero. Independent γ values (grey) extracted from the CW Rabi data showing no clear dependence on excitation power. The light grey bar indicates the expected value of the decoherence rate $\gamma = \gamma_2/2 = 0.78 \pm 0.03$ ns^{-1}, where the bar's width represents the error (taken from the exponential fit in Fig. 2). The error weighted mean is $\gamma = 0.82 \pm 0.04$ ns^{-1} with a standard deviation of 0.10 ns^{-1}.

though the frequency domain response of the system exhibits this spectral broadening, a time averaged measurement of the damped Rabi oscillations yields a decoherence rate, which is consistent with the theoretical limitation from the trion decay rate. This result is consistent with nonlinear measurements showing that the decoherence rate in InAs QDs is limited by the radiative lifetime,[23] a result that impacts the goal of scalable spin based QD network mediated by a spin-photon interface.[18,24]

In many scalable quantum computing architectures, the ability to coherently map information between distant qubits is crucial,[24–26] and is limited by the optical decoherence rate.[27,28] Specifically, this coherence is required in quantum information protocols that rely on coherent mapping between a single electron spin qubit confined to a QD and a photonic qubit.[24] This coherent spin-photon interface can be used to deterministically entangle spin qubits through an appropriately designed optical cavity.[24] The long optical coherence time, extracted from these data, is also promising for probabilistic QD entanglement schemes, which rely on interference between spontaneously emitted photons from distinct QDs.[29]

By extending the temporal duration of the driving pulse, we can measure the coherent transient response of the resonance fluorescence following step function excitation to directly observe the time-dependent Rabi oscillations of a two-level QD system. By direct detection of the Rabi oscillations using this time correlated single-photon technique, we are able to obtain data sets in two minutes that are of sufficient quality to extract the decoherence rate of the optical transition and directly measure the Rabi frequency of an applied pulse. Our time gated resonance fluorescence technique provides a time selective spin readout channel which is crucial for applications using QD spins as qubits. In summary, by using the coherent transient response of the resonance fluorescence, we find that $\gamma \approx \gamma_2/2$, indicating the absence of pure dephasing (i.e., decoherence due to purely phase destroying interactions), a result that is important in quantum information applications of single InAs QDs.

ACKNOWLEDGMENTS

The authors would like to acknowledge useful discussion with P. Berman, L. Webster, B. Sun, C. Chow, V. Lal, and K. Truex. This research is supported by NSF PHY0804114 and PHY1104446, AFOSR FA9550- 09-1-0457, DARPA FA9550-10-1-0534 and FA8750-12-2-0333, ARO W911NF-08-1-0487 and MURI W911NF-09-1-0406.

*dst@umich.edu

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